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(54) TERPOLYMERS AND HIGHER POLYMERS OF N-ALKYL-OR N-ALKOXYALKYL-ACRYLAMIDES OR -METHACRYLAMIDES, PROCESS FOR THEIR PREPARATION, AND THEIR USE IN COSMETICS

We, L'OREAL, a French Body Corporate, of 14 Rue Royale 75008, Paris, France, do hereby declare the invention for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:-

The present invention relates to terpolymers and higher copolymers based on N-alkyl- or N-alkoxyalkyl-acrylamide or N-alkyl- or N-alkoxyalkyl methacrylamide and their use in cosmetics, especially in lacquers and wavesetting lotions.

The present invention also relates to a process for the preparation of these copolymers.

A large number of synthetic polymers have already been proposed for use in lacquers or wavesetting lotions.

It has now been found that it is possible to produce excellent lacquers and wavesetting lotions using the copolymers of this invention.

The copolymers according to the invention, contrary to those previously used, impart excellent properties to the lacquers and wavesetting lotions and in particular provide a good hold for a hairstyle.

Accordingly, the present invention provides copolymers, and especially terpolymers, containing units resulting from the copolymerisation of solely:

(a) 5 to 90% by weight of at least one monomer of the formula:

$$CH_{2} = C - C - NH - C - (X)_{a} - R$$

$$0 R_{a}$$
(I) 20

in which:

R represents a linear or branched alkyl radical having from 1 to 10 carbon atoms,  $R_1$ ,  $\tilde{R}_2$  and  $R_3$  each represents a hydrogen atom or a methyl radical, n is 0 or 1, and, if n=1, X represents an oxygen atom;

(b) 5 to 90% by weight of at least one monomer of the formula:

$$R_4 - CH = C - (CH_2)_m - CONH_2$$

$$R_4 - CH = C - (CH_2)_m - CONH_2$$

$$R_4 - CH = C - (CH_2)_m - CONH_2$$

$$R_4 - CH = C - (CH_2)_m - CONH_2$$

in which:

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m is 0 or 1,

-COO (ammonium group);

and, if m = 0, R4 represents a hydrogen atom, -COOH or -COO (ammonium

group) and R<sub>s</sub> represents a hydrogen atom or a methyl radical, or, if m = 1, R4 represents a hydrogen atom and R5 represents -COOH or 5

and (c) 5 to 60% by weight of at least one monomer which is styrene giving rise to units of formula

N-vinylpyrrolidone giving rise to units of formula

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or a monomer of any one of the following formulae:

$$CH_2 = C - CN$$

$$R_a$$
(III)

giving rise to units of formula

$$\begin{bmatrix} CN & \\ CH_2 & C \\ \\ R_6 \end{bmatrix}$$
 (IIIa)

15 in which:

R<sub>6</sub> represents a hydrogen atom or a methyl radical:

(IV)

giving rise to units of formula

20 in which:

20  $\mathbf{R}_{7}$  represents a hydrogen atom or a methyl radical and

R<sub>s</sub> represents a linear or branched alkyl radical having from 1 to 18 carbon atoms, a quaternised or non-quaternised

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a ---CH2CH2OH radical,

or a -(CH2-CH2O)1R' radical,

in which R' represents a methyl or ethyl radical and 1 is 12;

 $\begin{array}{ccc}
R_s - C - O - CH = CH_z & (V) \\
\parallel & O
\end{array}$ 

giving rise to units of formula

$$\begin{bmatrix}
R_9 \\
C=0 \\
O \\
CH-CH_2
\end{bmatrix}$$
(Va)

in which:

 $\overline{R}_{o}$  represents a linear or branched alkyl radical having from 1 to 16 carbon atoms;

CH\_COOR"

(VI)

giving rise to units of formula

in which: .

R" represents an alkyl radical having from 1 to 3 carbon atoms;

----

$$CH_2 = CH - O - R_{10}$$
 (VII)

giving rise to units of formula

$$\begin{array}{c|c}
 & & & R_{10} \\
 & & & & O \\
 & & & & & CH_2
\end{array}$$
(VIIa)

in which:

 $R_{10}$  represents a linear or branched alkyl radical having from 1 to 17 carbon atoms; and

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$$R_{13}$$
— $CH = C$ — $(CH_2)_p$ — $CON$ — $Z$ — $OH$  (VIII)

giving rise to units of formula

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$$\begin{array}{c|c}
 & R_{11} \\
 & N - Z - OH \\
 & CO \\
 & (CH_2)_p \\
 & CH - C \\
 & R_{13} & R_{12}
\end{array}$$
(VIIa)

in which:

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R<sub>11</sub> represents a hydrogen atom or a methyl radical,

Z represents a linear or branched alkylene radical having 1 to 6 carbon atoms, which may or may not be substituted by one or two hydroxymethyl groups, p is 0 or 1,

and, if p=0, R<sub>13</sub> represents a hydrogen atom or -COR<sub>14</sub>, in which R<sub>14</sub> repre-

sents -OH, -O (ammonium group) or -NH-R<sub>15</sub>, in which R<sub>15</sub> represents a hydrogen atom or -Z-OH, and R<sub>12</sub> represents a hydrogen atom or

the —CH<sub>3</sub> radical, or, if p=1,  $R_{13}$  represents a hydrogen atom and  $R_{12}$  represents —COR<sub>14</sub>,  $R_{14}$ 

having the same meaning as above.

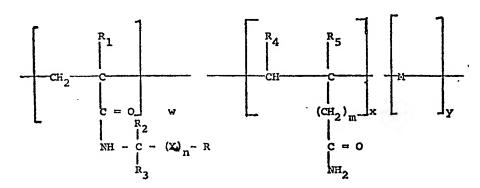
It will be appreciated that the expression "ammonium group" covers not only NH4 but also substituted ammonium groups.

In the formula (VIII) above, the radical Z preferably represents:

$$\begin{array}{c} -CH_2-, \\ -(CH_2)_2-, \\ -CH(C_2H_5)-CH_2-, \\ -C(CH_3)_2-CH_2-, \\ -C(CH_2OH)_2-CH_2-, \\ -C(CH_3)_2-CH_2-CH(CH_3)-, \\ \text{or } -C(CH_3)(CH_2OH)-CH_2-. \end{array}$$

As indicated above, the copolymers according to the invention are preferably terpolymers. However, the copolymers can be tetrapolymers or pentapolymers or higher copolymers. In the case of tetrapolymers or higher copolymers, the copolymers result from the copolymerisation of more than one monomer of the formula (I) and/or more than one monomer of the formula (II) and/or more than one of the monomers represented by the above formulae (III) to (VIII).

30 The terpolymers according to the invention can be represented by the following general formula:



in which: R, R<sub>1</sub>, R<sub>2</sub>, R<sub>3</sub>, R<sub>4</sub> and R<sub>5</sub>, n, m and X are as defined above for the formulae (I) and (II), w corresponds to 5 to 90% by weight, x corresponds to 5 to 90% by weight and y corresponds to 5 to 60% by weight, and M represents a unit derived from styrene, N-vinylpyrrolidone or a monomer of any one of the formulae (III) to (VIII) given above.

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	Examples of monomers of the formula (I), include particularly N-tertiary butyl-	
	acrylamide, N-octylacrylamide, N-decylacrylamide, N-dodecylacrylamide, N-[(1.1-	
	dimethyl) - propyl - 1] - acrylamide, N - [(1,1 - dimethyl) - butyl - 1] - acrylamide,	
E	N - [(1,1 - dimethyl) - pentyl - 1] - acrylamide, N isobutoxymethylacrylamide as	_
5	well as the corresponding methacrylamides.	5
	Examples of monomers of the formula (II), include particularly acrylamide,	
	methacrylamide, maleamic acid and itaconamic acid.	
	Examples of monomers of the formula (III) to (VIII), include, particularly,	
10	acrylonitrile, methacrylonitrile, methyl, ethyl, propyl, isopropyl, butyl, tertiary butyl,	10
ľ0	hexyl, decyl, dodecyl, octadecyl, 2-hydroxyethyl and quaternised or non-quaternised	10
	2-N.N-dimethylaminoethyl acrylate and methacrylate, and w-methyl- or w-ethyl-	
	polyethylene glycol acrylate and methacrylate; vinyl acetate, vinyl propionate, vinyl	
	butyrate, vinyl laurate, vinyl stearate, vinyl pivalate, vinyl neoheptanoate, vinyl neo-	
15	octanoate, vinyl neodecanoate, vinyl 2,2,4,4 - tetramethyl - valerate and vinyl 2 - iso-	15
IJ	pronyl - 2,3 - dimethyl - butyrate; dimethyl maleate, diethyl maleate, dimethyl itacon-	15
	ate and diethyl itaconate; methyl vinyl ether, ethyl vinyl ether, butyl vinyl ether, iso-	
	propyl vinyl ether, octyl vinyl ether, dodecyl vinyl ether and octadecyl vinyl ether;	
	N - hydroxymethylacrylamide, N - hydroxymethylmethacrylamide, N - (2 - hydroxy-	
20	ethyl) - acrylamide, N - (2 - hydroxyethyl) - methacrylamide, N - hydroxymethyl-	20
	maleamic acid, N - hydroxymethyl - maleamide, N,N' - dihydroxymethyl - maleamide,	
	N - hydroxymethyl - itaconamic acid, N - hydroxymethyl - itaconamide, N,N - di-	
	hydroxymethyl - itaconamide, N - [(1 - hydroxymethyl) - propyl] - acrylamide, N-	
	[(1 - hydroxymethyl) - propyl] - methacrylamide, N - [(1 - methyl - 1 - hydroxymethyl) - ethyl - 1] - oprologide N [(1 - methyl - 1 - hydroxymethyl) - ethyl - 1]	
25	methyl) - ethyl - 1] - acrylamide, N - [(1 - methyl - 1 - hydroxymethyl) - ethyl - 1]-	25
	methacrylamide, N - [1,1,1 - tris - (hydroxymethyl) - methyl] - acrylamide, N- [1,1,1 - tris - (hydroxymethyl) - methyl] - methacrylamide, N - [(3 - hydroxy - 1,1-	
	dimethyl) - butyl] - acrylamide, N - [(3 - hydroxy - 1,1 - dimethyl) - butyl]-	
	methacrylamide, N - (2 - hydroxyethyl) - N - methyl - acrylamide, N - (2 - hydroxy-	
	ethyl) - N - methyl - methacrylamide, N - [1,1 - bis - (hydroxymethyl) - ethyl]-	20
30	acrylamide and N - [1,1 - bis - (hydroxymethyl) - ethyl] - methacrylamide.	30
	The copolymers according to the invention preferably have a molecular weight of	
	1,000 to 500,000 and more particularly a molecular weight of 2,000 to 200,000.	
	In a particular embodiment the copolymers according to the invention are cross-	
25	linked with a crosslinking agent used in a proportion of 0.01 to 2% by weight based	35
35	on the total weight of the monomers employed for the reaction.	
	Examples of various crosslinking agents which can be used, include particularly	
	diethylene glycol dimethacrylate, diallyl ether, tetraallyloxyethane, ethylene glycol	
	dimethacrylate and ethylene glycol diacrylate.	
40	According to a particular embodiment, if the radical R, represents a free carb-	40
	oxylic acid group, the latter can be neutralised with at least one organic base, such	
	as monoethanolamine, diethanolamine, triethanolamine, the isopropanolamines such as	
	triisopropanolamine, morpholine as well as certain amino-alcohols such as 2-amino-2-	
	methyl-propanol and 2-amino-2-methyl-1,3-propanediol.	4-
45	The carboxyl groups can suitably be neutralised with these organic bases in a proportion of 10 to 150%.	45
	The present invention also relates to a process for the preparation of copolymer	
	such as those described above.	
	These copolymers can be prepared by solution copolymerisation in an organic	
50	solvent such as an alcohol, an ester, a ketone or a hydrocarbon.	50
50	Examples of solvents include particularly methanol, isopropanol, ethanol, ethyl	
	acetate, ethyl methyl ketone, and benzene.	
	The copolymerisation can also take place in suspension or in emulsion in an	
	mert solvent such as water.	
55	The copolymerisation can also take place in bulk.	55
	These copolymerisations can be carried out in the presence of a polymerisation	
	catalyst which generates free radicals, such as benzoyl peroxide, lauroyl peroxide,	
	azo-bis-isobutyronitrile, hydrogen peroxide and various oxidation-reduction combina-	
60	tions such as (NH <sub>4</sub> ) <sub>4</sub> S <sub>2</sub> O <sub>8</sub> with FeCl <sub>2</sub> .  The caralyst concentration is switchly 0.2 to 10% by mainly bound on the mine.	۲۸
60	The catalyst concentration is suitably 0.2 to 10% by weight based on the weight of the monomers used for the reaction and in accordance with the molecular weight	60
	of the copolymers which it is desired to obtain.	
	The present invention furthermore relates to the use of the copolymers of the	
	invention in cosmetic compositions.	
	<b>▲</b> • • • • • • • • • • • • • • • • • • •	

6	1,572,626	6
	In particular, the present invention relates to cosmetic compositions which are in the form of lacquers or wavesetting lotions.  These cosmetic compositions contain at least one copolymer of this invention in	
,5	solution in an appropriate cosmetic vehicle.  The cosmetic compositions according to the invention can be, for example, hair lacquers which may or may not be in the form of an aerosol, wavesetting lotions, hair	5
10	treatment compositions, dyeing carriers, shampoos or compositions called "rinses" which are applied to the hair after washing the head of hair with a shampoo.  By way of example, a hair lacquer aerosol can be produced by packaging, in an	
10	aerosol container, 0.2 to 8% by weight of a copolymer according to the invention and 6 to 30%, preferably 8 to 25%, by weight of an alcohol, and a propellant gas liquefied under pressure, such as dichlorodifluoromethane, trichlorofluoromethane, nitrous oxide or carbon dioxide or mixtures of these.	10
15	Preferably, ethanol or isopropanol is used as the alcohol.  The wavesetting lotions according to the invention can be produced, for example, by introducing 0.3 to 6% by weight of a copolymer according to the invention into an aqueous-alcoholic solution, preferably having an alcohol content of 20 to 66%.  The cosmetic compositions according to the invention can also contain conven-	15
20	tional cosmetic adjuvants such as perfumes, dyestuffs, preservatives, plasticisers, cationic products, non-ionic products, silicones for improving the gloss, or other cosmetic resins.	20
	The following Examples of the preparation of the copolymers and of compositions based on them serve to illustrate the invention.	
	Examples of the Preparation of Copolymers	
25	EXAMPLE 1.  400 g of ethanol, 55 g of N-tertiary butylacrylamide, 27.5 g of acrylamide, 17.5 g of methylmethacrylate and 1 g of azo-bis-isobutyronitrile are introduced into a one	25
30	litre flask equipped with a condenser, a mechanical stirrer and a nitrogen inlet tube.  The reaction mixture is heated at 80°C for 8 hours by means of a thermostatically controlled apparatus and is then allowed to cool to ambient temperature.  The solution is then poured dropwise into a vessel containing ethyl acetate as a precipitors.	30
	precipitant. The precipitated polymer is then filtered off and dried under reduced pressure.  Yield: 76%.	
35	Viscosity: 2.95 cPo (as a 5% strength solution in DMF at 34.6°C).	35
	EXAMPLE 2.  65 g of N-tertiary butylacrylamide, 20 g of acrylamide, 15 g of N-hydroxymethylacrylamide and 1 g of azo-bis-isobutyronitrile are copolymerised using the method described in Example 1.	
40	Viscosity: 2.61 cPo (5% strength solution in DMF at 34.6°C).  Other Examples of copolymers (Examples 3 to 17) are given in Table I, the quantities are expressed in grams. All these copolymers were prepared in accordance with Example 1, only the nature of the precipitant being different.	40

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N.:mples	~	-1	5	\[ \]	7	CS	6	92	Ξ	12	13	14	15	92	17
N-T-Butylacrylamide	50	9	50	09	8	50	35	\$	8	\$	07	30	20	35	92
Acrylamide	25	20	30	30	15	35	15	40	40	94	20	30	55	25	5
N.Vinylpyrrolidone	25	40												30	
Acrylonitrile			20		,										
Styrene				10											
2-Hydroxyethyl Methacrylate					25										
Methyl Methacrylate						15	10								25
Steary! Methacrylate							5								
Vinyl Acetate								70			•				
Diethyl Maleate									20						
Butyl Vinyl Ether										30					
Dimethylaminoethyl Methacrylate							35				<del>\$</del>				
Polyethylene Glycol Monomethyl Ether Methacrylate												40		20	9
N-[(1-Methyl-1-Hydroxymethyl)- Ethyl-1]-Acrylamide													25		
Precipitant	0	0	0	0	0	0	<u>(2)</u>	0	0	0	0	<b>a</b>	<u>©</u>	0	Θ
Viscosity (at 5% strength in DMF), 34.6°C in cPo	2.22	2.32	2.00	2,10	2.18	2.54	2.31	1.85	1.60	2.15	2.05	2.90	2.35	2.18	2.18

Ref.: (1) Petroleum ether (2) Diethyl ether (3) Acetone (4) Heptane

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# Examples of Compositions

EXAMPLE A. A wavesetting lotion is prepared, according to the invention, by mixing the following ingredients

5 Polymer prepared according to Example 1 Perfume 0.1 g Ethanol 45 100 Water, q.s.p.

This wavesetting lotion, applied in the conventional manner, makes it possible to 10 impart to the hair a glossy appearance, and excellent hold over a period of time. In this Example, the copolymer prepared according to Example 1 can be replaced 10 by an equal amount of the copolymer prepared according to Example 5, 10-12 or 15.

EXAMPLE B. A hair lacquer is prepared, according to the invention, by mixing the following 15 ingredients:

> Polymer prepared according to Example 2 Perfume 0.3 g Ethanol, q.s.p. 100 g

25 g of this solution are packaged in an aerosol container with 47 g of trichloro-20 fluoromethane and 28 g of dichlorodifluoromethane. 20 By spraying this lacquer, an attractive glossy appearance of the head of hair, and

a high lacquering power are achieved.

The hair is soft to the touch and the lacquer is easily removed by combing or brushing.

25 In this Example, the copolymer prepared according to Example 2 can be replaced 25 by an equal amount of copolymer prepared according to Example 3, 4, 6-8 or 10.

#### EXAMPLE C.

An aerosol hair lacquer is prepared, according to the invention, by mixing the following ingredients:

30 Polymer prepared according to Example 11 5 g 0.07 g 100 g 30 Perfume Ethanol, q.s.p.

93 g of this solution are then packaged in an aerosol container and carbon dioxide is introduced so as to bring the internal pressure to about 8 bars.

35 In this Example the polymer prepared according to Example 11 can advantageously be replaced by the same amount of one of the polymers prepared according to Example 12, 14 or 16.

#### EXAMPLE D.

A wavesetting lotion is prepared, according to the invention, by mixing the 40 following ingredients:

Polymer prepared according to Example 9 2 (quaternised with dimethyl sulphate) Perfume 0.1 g Isopropanol 45 g 45 Water, q.s.p. 45

This wavesetting lotion, applied in the conventional manner, makes it possible to impart to the hair a glossy appearance, and excellent hold over a period of time. The polymer according to Example 9 can advantageously be replaced by the same amount of the polymer according to Example 13, which has also been quaternised.

#### EXAMPLE E.

A hair lacquer is prepared, according to the invention, by mixing the following ingredients:

5

Polymer prepared according to Example 2 Perfume Ethanol

4 g 0.3 g

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25 g of this solution are packaged in an aerosol container with 47 g of trichlorofluoromethane and 28 g of dichlorodifluoromethane. By spraying this lacquer, an attractive glossy appearance of the head of hair, and

a high lacquering power are achieved.

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The hair is soft to the touch and the lacquer is easily removed by combing or

In this Example, the polymer according to Example 2 can advantageously be brushing. replaced by the same amount of the polymer according to Example 17.

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WHAT WE CLAIM IS:-

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1. A copolymer containing units derived solely from:

(a) 5 to 90% by weight of at least one monomer of formula:

$$\begin{array}{c|c}
R_1 & R_2 \\
\downarrow & \downarrow \\
CH_2 = C - C - NH - C - (X)_n - R \\
\downarrow & \downarrow \\
O & R_3
\end{array}$$
(I)

in which:

R represents a linear or branched alkyl radical having up to 10 carbon atoms, R<sub>1</sub>, R<sub>2</sub> and R<sub>3</sub> each independently represents a hydrogen atom or a methyl radical, n is 0 or 1

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and X represents an oxygen atom;

(b) 5 to 90% by weight of at least one monomer of formula:

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ight of at least the monotone 
$$R_4$$
— $CH=C$ — $(CH_2)_m$ — $CONH_2$  (II) 25
 $R_4$ 

in which

if m = 0,  $R_4$  represents a hydrogen atom, —COOH or —COO (ammonium

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and (c) 5 to 60% by weight of at least one of styrene, N-vinylpyrrolidone and a monomer of any one of the formulae:

$$CH_2 = C - CN$$

$$R_6$$
(III)

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 $R_e$  represents a hydrogen atom or a methyl radical:

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in which:

R<sub>7</sub> represents a hydrogen atom or a methyl radical and

Ra represents a linear or branched alkyl radical having up to 18 carbon atoms, a —(CH<sub>2</sub>)<sub>2</sub>—N(CH<sub>3</sub>)<sub>2</sub> radical which may be quaternised, a —CH<sub>2</sub>CH<sub>2</sub>OH radical,

45

or a —(CH2—CH2O)lR' radical, in which R' represents a methyl or ethyl radical

$$\begin{array}{ccc}
R_{\circ}-C-O-CH=CH_{2} \\
\emptyset \\
O
\end{array} (V)$$

5 in which:

R, represents a linear or branched alkyl radical having up to 16 carbon atoms;

CH-COOR" ĊH—COOR" (VI)

in which:

R" represents an alkyl radical having from 1 to 3 carbon atoms;

 $CH_2 = CH - O - R_{1c}$ in which: (VII) 10

R<sub>10</sub> represents a linear or branched alkyl radical having up to 17 carbon atoms;

$$R_{13}$$
— $CH = C$ — $(CH_2)_p$ — $CON$ — $Z$ — $OH$  (VIII)

15 in which:

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R<sub>11</sub> represents a hydrogen atom or a methyl radical, 15

Z represents a linear or branched alkylene radical having up to 6 carbon atoms, which is unsubstituted or substituted by one or two hydroxymethyl groups,

p is 0 or 1 such that if p=0, R<sub>13</sub> represents a hydrogen atom or —COR<sub>14</sub> in which R<sub>14</sub> represents 20

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—OH, —O (ammonium group) or —NH—R<sub>15</sub>, in which R<sub>16</sub> represents a hydrogen atom or —Z—OH, and R<sub>12</sub> represents a hydrogen atom or a methyl

or, if p=1,  $R_{13}$  is a hydrogen atom and  $R_{12}$  is  $COR_{14}$  in which  $R_{14}$  is as defined

25 2. A copolymer according to claim 1 which has a molecular weight of 1,000 to 500,000. 25

3. A copolymer according to claim 2 which has a molecular weight of 2,000 to 200,000.

30 4. A copolymer according to any one of the preceding claims which is a terpolymer.

5. A copolymer according to any one of claims 1 to 3 which is crosslinked with a crosslinking agent used in a proportion of 0.01 to 2% by weight based on the total

35 6. A copolymer according to claim 5 in which the crosslinking agent is diethylene glycol dimethacrylate, diallyl ether, tetraallyloxyethane, ethylene glycol dimethacrylate

35 7. A copolymer according to any one of the preceding claims in which the monomer of formula (I) is N-tertiary butylacrylamide, N-octylacrylamide, N-decylacrylamide, N-dodecylacrylamide, N - [(1,1 - dimethyl) - propyl - 1] - acrylamide, N - [(1,1 - dimethyl) - butyl - 1] - acrylamide, N - [(1,1 - dimethyl) - pentyl - 1]acrylamide or a corresponding methacrylamide. 40

8. A copolymer according to any one of the preceding claims in which the monomer of formula (II) is acrylamide, methacrylamide, maleamic acid or itaconamic

9. A copolymer according to any one of the preceding claims in which the monomer of any one of the formulae (III) to (VIII) is acrylonitrile, methacrylonitrile, methyl, ethyl, propyl, isopropyl, butyl, tertiary butyl, hexyl, decyl, dodecyl, octadecyl,

25. A composition according to claim 18 substantially as described in any one

of Examples A to E.

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